

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicants: Masaru ISHINO et al.
Title: METHOD FOR PRODUCING PROPYLENE OXIDE
Appl. No.: 10/593,267
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Examiner: David E. Gallis
Art Unit: 1625
Confirmation Number: 6446

DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Madam/Sir:

I, the undersigned, Hiroaki ABEKAWA, a citizen of Japan, residing at 9-6-5-2306 Honmachi, Toyonaka-shi, Osaka, Japan declare and state:

I completed my Bachelor's degree in the Department of Organic and Polymeric Materials at the Tokyo Institute of Technology, Faculty of Engineering, in March, 1992.

I joined Research Laboratories of Sumitomo Chemical Company, Limited, and was engaged in the research of Catalyst for Organic or Inorganic Synthesis from April, 1992 to present.

I am an inventor of the above-identified application and am familiar with the subject matter thereof. I have studied the Office Action.

The Examiner has alleged that because the disclosure is enabling for only the use of hydrogen peroxide as the peroxide and tert-butyl alcohol as the solvent, the "specification does not enable any person skilled in the art to use the invention commensurate in scope with these claims." The following experiments were made under my supervision in order to show that the claimed invention and specification fulfills the requirements under 35 USC § 112. The experiments show that with the same methods recited in the specification, different alcohols, in addition to tert-butyl alcohol, can be used.

1) Preparation of Titanosilicate Catalyst

The titanisilicate precursor used in the following experiments were prepared by the following procedures:

A gel was prepared by dissolving 256g of piperidine, 687g of pure water, 161g of boric acid, 32g of TBOT (tetra-n-butyl orthotitanate) and 119g of fumed silica (cab-o-sil M7D) under stirring in an autoclave at room temperature in 1 atm, and aged for 1.5 hours, and thereafter tightly closed. The gel was heated-up for over 8 hours while stirred, and thereafter kept at 160°C for 120 hours to obtain a suspension by hydrothermal synthesis. The obtained suspension was filtered and thereafter washed in water until the filtrate had a pH of about 10. Next, the filter cake was dried at 50°C to obtain wet white powder. 750 mL of 13% by weight-nitric acid was added to 15g of the obtained powder and refluxed for 20 hours. Subsequently, the powder was filtered, washed with water to the proximity of neutrality and sufficiently dried at 50°C to obtain 22g of wet white powder, containing 40% water by weight. The white powder was subjected to an X-ray diffraction pattern analysis using an X-ray diffractometer with copper K- α radiation, which showed the x-ray diffraction pattern in Fig. 1. Thus, the catalyst to be used in the present invention was obtained. It was confirmed that the obtained white powder had 1.1wt% Ti by ICP emission analysis. The catalyst is hereinafter referred to as Cat. A.

2 θ /°	Lattice spacing d/Å
6.47	13.6
7.33	12.1
8.05	11.0
9.71	9.1
13.0	6.8
22.8	3.9
25.4	3.5
26.3	3.4

- 2) Epoxidation reaction of propylene using Cat. A.

Experiment 1

The reaction was performed by using Cat A.

An aqueous 5wt% H₂O₂ solution was prepared by mixing 7g of 30% H₂O₂ (Wako Pure Chemicals Industries, Ltd), 20g of tert-butanol (Wako Pure Chemicals Industries, Ltd) and 15g of pure water. 12 g of the prepared solution and 0.010 g of the pulverized Cat. A were charged into a 50-ml stainless-steel autoclave. Next, the autoclave was moved to an ice bath, which was filled with 10g of liquefied propylene and further pressurized up to 2MPa-G with Argon. The autoclave was put in a hot water bath until the internal temperature became 40°C, and the time 5 minutes after when the internal temperature rose up to approximately 40°C was regarded as reaction initiation. 1 hour after the reaction initiation, the autoclave was taken out of the hot water bath for sampling. The pressure after initiation of the reaction was 2.8 MPa-G. The reaction mixture was analyzed by using gas chromatography. As a result, propylene oxide production activity per unit catalyst weight was 0.687 mol·h⁻¹·g⁻¹.

Experiment 2

An experiment was conducted in a similar manner to Experiment 1, except that 20g of 2-propanol (Nacalai Tesque Inc.) was used in place of tert-butanol, and 20g of the aqueous 5wt%

H₂O₂ solution was used. As a result, propylene oxide production activity per unit catalyst weight was 0.399 mol·h⁻¹·g⁻¹.

Experiment 3

An experiment was conducted in a similar manner to Experiment 1, except that 20g of methanol (Wako Pure Chemical Industries, Ltd) was used in place of tert-butanol, and 12g of an aqueous 5wt% H₂O₂ solution was used. As a result, propylene oxide production activity per unit catalyst weight was 0.462 mol·h⁻¹·g⁻¹.

Experiment 4

An experiment was conducted in a similar manner to Experiment 1, except that 20g of acetone (Kanto Chemical Co., Inc.) was used in place of tert-butanol, and 12g of an aqueous 5wt% H₂O₂ solution was used. As a result, propylene oxide production activity per unit catalyst weight was 0.629 mol·h⁻¹·g⁻¹.

3) Results and conclusions:

Table 1 below shows propylene oxide formation reaction results using tert-butanol or acetonitrile solvent of the present specification.

Table 2 below shows propylene oxide formation reaction results using tert-butanol, 2-propanol, methanol or acetone solvent.

Therefore, from the experimental results shown below, one of ordinary skill in the art will understand that 2-propanol, methanol, and acetone, which are disclosed in the present specification, are effective solvents for the epoxidation reaction of propylene using a titanasilicate precursor as a catalyst in a similar reaction operation as disclosed for tert-butanol or acetonitrile.

Table 1.

	Exp. 1	Exp. 2
Catalyst	Ti-MWW precursor	Ti-MWW precursor
Solvent	t-Butanol /water	Acetonitrile /water
activity* mol·g-cat·h ⁻¹	0.781	0.319

*activity: propylene oxide production activity per unit catalyst weight

Table 2

	Exp. 1	Exp. 2	Exp. 3	Exp. 4
Catalyst	Cat. A	Cat. A	Cat. A	Cat. A
Solvent	t-Butanol /water	2-propanol /water	Methanol /water	Acetone /water
activity* mol·g-cat·h ⁻¹	0.687	0.399	0.462	0.629

*activity: propylene oxide production activity per unit catalyst weight

I further declare that all the statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further, that these statements are made with the knowledge that willful false statements so made are punishable by fine or imprisonment, or both, under Section 101 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: June 24, 2008

By: Hiroaki ABEKAWA

Hiroaki, ABEKAWA

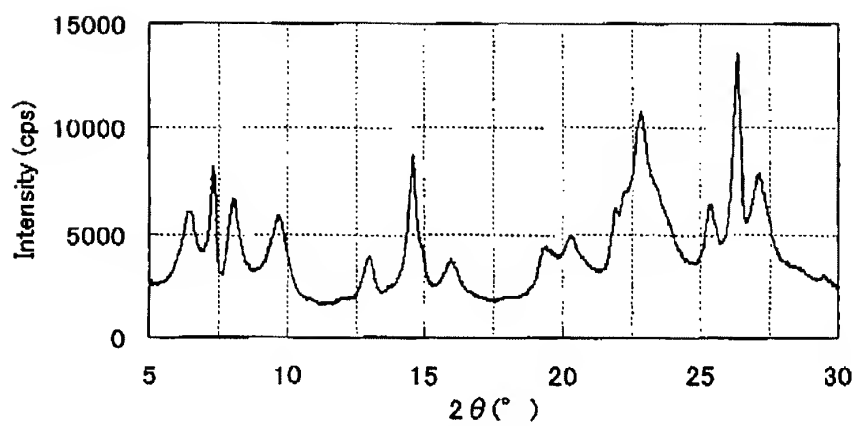


Figure 1